# **Viscosity Measurements on Nitrogen**

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The viscosity coefficient of gaseous nitrogen was measured with a vibrating-wire viscometer of very high precision. The measurements were performed along the six isotherms of (298.15, 323.15, 348.15, 373.15, 398.15, and 423.15) K and at pressures up to a maximum of 35 MPa. The gas densities needed for the evaluation of the measuring values were calculated using an equation of state by Span et al. (*J. Phys. Chem. Ref. Data* **2000**, *29*, 1361–1433). The reproducibility is  $\pm$  0.05 %, whereas the total uncertainty is estimated to be  $\pm$  (0.2 to 0.3) %. The viscosity values of the isotherms were evaluated with a power series expansion in terms of reduced density. A comparison with the most recent correlation by Lemmon and Jacobsen (*Int. J. Thermophys.* **2004**, *25*, 21–69) shows that the new values agree in the temperature and density ranges of the measurements within  $\pm$  0.7 %. Based on a comparison with the most reliable data from the literature, it is concluded that the new values are appropriate to improve the viscosity surface correlation of nitrogen.

## Introduction

Air is one of the working fluids mostly used in industrial applications, such as in combustion processes, compressors, and turbines. Unfortunately, the thermophysical properties of air have not been investigated very often and with sufficiently high accuracy. The reason for that is the risk of measurements on oxygen or on mixtures with a high content of oxygen. On the contrary, the thermophysical properties of nitrogen as the main component of air have been measured rather frequently with high accuracy. The most recent reference equation of state for the thermodynamic properties of nitrogen was reported by Span et al.<sup>1</sup> The uncertainty in the density using this equation is estimated to be less than  $\pm$  0.05 % in the temperature and pressure ranges of the viscosity measurements of the present paper.

The transport properties of nitrogen have not been determined with the same high accuracy as the thermodynamic data. Stephan et al.,<sup>2</sup> Millat and Vesovic,<sup>3</sup> and Lemmon and Jacobsen<sup>4</sup> presented transport property surface correlations for nitrogen in the complete fluid ranges. Whereas Millat and Vesovic did not give detailed information about the uncertainty of their correlation, Stephan et al. reported standard deviations for the viscosity of  $\pm$  0.3 % in the dilute-gas region and of  $\pm$  1 % for densities up to 18 mol·dm<sup>-3</sup> considering only a limited number of data sets from literature. The most recent correlation by Lemmon and Jacobsen is based on an extended database resulting in comparably higher uncertainties of the viscosity:  $\pm$  0.5 % in the dilute-gas region and  $\pm$  2 % in the temperature range of (270 to 370) K up to pressures of 100 MPa as well as higher values at the extremes of the fluid range. Nevertheless, this correlation will be used as the basis for a comparison, since further and new results of viscosity measurements have been taken into account as compared with

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the older correlations. With regard to the development of a high standard for the test and implementation of instruments for viscosity measurements on gases, the uncertainties of the mentioned correlations are still too large and should be further improved. The new measurements of the viscosity of nitrogen are intended to provide high-precision values in the temperature range of (298 to 423) K and up to moderate pressures of 35 MPa.

Some years ago Wilhelm et al.<sup>5</sup> designed a vibrating-wire viscometer and performed high-precision measurements on argon, krypton, and propane<sup>6,7</sup> and, recently, on sulfur hexafluoride.<sup>8</sup> A special measuring program was arranged to determine the viscosity of natural gas components and of natural gas mixtures to fulfill the needs of the gas industry. In addition to the investigation on propane, the viscosity of methane,<sup>9,10</sup> of two natural gases,<sup>10,11</sup> and very recently of ethane<sup>12</sup> were determined. The measurements on nitrogen reported in this paper complement this program.

# **Experimental Section**

A detailed description of the design and implementation of the vibrating-wire viscometer has been given by Wilhelm et al.5 and Wilhelm and Vogel<sup>6</sup> so that only some essential items should be summarized here. The wire with a length of 90 mm and a diameter of about 25  $\mu$ m is placed in a magnetic field in such a manner that all even and the third harmonic modes are suppressed. Chromel has been selected as wire material due to its comparably smooth surface. The oscillation is initiated by a sinusoidal voltage pulse with a frequency close to the resonant frequency of the wire. The oscillation following the pulse is detected by amplifying the induced voltage and measuring it as a function of time. One hundred oscillation curves are recorded and averaged to improve the signal-to-noise ratio. Comparably large displacements contrary to the requirements of the measuring theory are applied in order to obtain reasonably large measuring signals. Finally, the logarithmic decrement ( $\Delta$ ) and the frequency  $(\omega)$  of the oscillation, determined for different values of the relative initial amplitude  $\epsilon = y_{\text{max}}/R (y_{\text{max}} - \text{initial})$ 

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wire displacement, R – wire radius), are deduced by an extrapolation of  $\epsilon^2 \rightarrow 0$ .

The viscometer cell is suspended from the top closure of a pressure vessel placed in a heat-pipe thermostat. The pressure is determined with four transmitters supplied by Digiquartz. Their working ranges of (0.69, 2.76, 13.8, and 41.4) MPa are characterized by uncertainties of  $\pm$  0.01 % of full scale. The temperature is obtained by means of a premium ITS-90 thermometer and a  $6^{1/2}$  digit multimeter with an uncertainty of  $\pm$  0.015 K.

The theory of the vibrating-wire viscometer<sup>13</sup> provides working equations for the calculation of the viscosity ( $\eta$ ) from the deduced values  $\Delta$  and  $\omega$ . For that purpose, the density ( $\rho$ ) of the fluid, the density ( $\rho_s$ ), and the radius (R) of the wire as well as the values  $\Delta_0$  and  $\omega_0$  in vacuo are needed. The wire radius (R) has been derived from viscosity measurements on argon at room temperature to be (12.770 ± 0.005)  $\mu$ m. In doing so, reference values by Kestin and Leidenfrost<sup>14</sup> were used assuming the density of the wire material chromel to be 8500 kg·m<sup>-3</sup> as given by the supplier.

#### **Measurements and Results**

The certified purity of nitrogen supplied by Messer Griesheim (Germany) was 99.999 %. The isotherms at (298.15, 323.15, 348.15, 373.15, 398.15, and 423.15) K cover a pressure range up to 35 MPa. The densities ( $\rho$ ) were calculated from the measured pressures (p) and temperatures (T) with the equation of state by Span et al.<sup>1</sup>

The isothermal series of measurements comprise about 50 data points apart from the 298 K isotherm. The individual points were not measured exactly at the nominal temperatures, but the deviations of the experimental temperatures were kept within  $\pm$  0.05 K for the first three isotherms and within  $\pm$  0.1 K for the higher ones. Hence, the experimental viscosity values were adjusted to the nominal temperature using a Taylor expansion restricted to a second power in temperature. In this procedure, the densities directly derived from the experiments and those for the isotherms are the same. Then the pressures at the nominal temperatures of the isotherms were recalculated from the densities. The results for all series of measurements are listed as triples of pressure, density, and viscosity in Table 1. The experimental values at pressures of about 0.1 MPa were controlled with regard to a possible influence of slip due to a comparably small density. This effect was observed in the measurements on propane,7 sulfur hexafluoride,8 and methane.10 But only one value in each series of measurements was found to be influenced by slip. These values are included, but marked in the table, and excluded from further evaluation.

The reproducibility of the measurements is better than  $\pm 0.05$  %. With regard to the absolute uncertainty, errors resulting from the calibration values by Kestin and Leidenfrost,<sup>14</sup> which are characterized by an uncertainty of  $\pm 0.1$  %, as well as from the density determination have to be considered above all. In general, an uncertainty of  $\pm 0.025$  % of the density data contributes a change of  $\pm 0.025$  % in the viscosity, whereas the allocation error related to temperature and pressure is not exceeding  $\pm 0.02$  %. As a result, the total uncertainty of the viscosity values is estimated to be about  $\pm 0.2$  %, possibly increasing up to  $\pm 0.3$  % at the highest measured pressures.

#### Analysis

The experimental results of the isotherms were correlated as a function of the reduced density ( $\delta$ ) by means of a power series



**Figure 1.** Evaluation of the 298.15 K isotherm with a power series expansion in the reduced density ( $\delta$ ) (eq 1). Representation of the weighted standard deviation ( $\sigma$ ) as a function of the maximum molar density ( $\rho_{max}$ ) for which experimental points are included.  $\bigcirc$ , first order in  $\delta$ ;  $\triangle$ , second order in  $\delta$ ;  $\diamondsuit$ , third order in  $\delta$ ;  $\bigtriangledown$ , fourth order in  $\delta$ .

representation restricted to the fourth or a lower power depending on the included density range.

$$\eta(\tau, \delta) = \sum_{i=0}^{n} \eta_i(\tau) \delta^i$$

$$\delta = \frac{\rho}{\rho_{c,N_2}} \quad \rho_{c,N_2} = 11.184 \text{ mol} \cdot \text{dm}^{-3}$$

$$\tau = \frac{T}{T_{c,N_2}} \quad T_{c,N_2} = 126.192 \text{ K}$$
(1)

The values of the critical constants correspond to that given by Span et al.<sup>1</sup> Weighting factors of  $(100/\eta)^2$  were used in the multiple linear least-squares regression to minimize the relative deviations. The weighted standard deviation ( $\sigma$ ), related to the maximum density of the experimental points included in the fit by means of eq 1, was used as criterion for the description of the considered isotherm.

Figure 1 shows that for the 298.15 K isotherm, which includes experimental values up to 11.8 mol·dm<sup>-3</sup>, a series expansion according to eq 1 with a fourth order in the reduced density ( $\delta$ ) is needed. For the higher isotherms a series expansion of either fourth or third or even second power is appropriate to describe the experimental points. The power in  $\delta$  of the series expansion needed for all six isotherms and the values of the coefficients ( $\eta_i$ ) are given in Table 2. Table 2 shows that the density coefficients ( $\eta_1$  to  $\eta_4$ ) seem to be only weakly temperature dependent.

With respect to a comparison with data from the literature discussed below, the experimental data of all isotherms were correlated with a double polynomial series expansion according to the following equation for which the coefficients  $\eta_{ij}$  are given in Table 3:

$$\eta(\tau, \delta) = \sum_{i=0}^{4} \sum_{j=0}^{2} \eta_{ij} \frac{\delta^{i}}{\tau^{j}}$$
(2)

Figure 2 illustrates that the deviations of the experimental values from the fit are practically within  $\pm 0.1$  for all isotherms. This

Table 1.	Experimental	Viscosity	Values of	Nitrogen	along	Isotherms
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$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		- F										
MPa         moldm <sup>-1</sup> μPa         MPa         moldm <sup>-2</sup> μPa         MPa         moldm <sup>-2</sup> μPa           53027         118.34         28.313         1.6010         6.2634         22.114         4.7899         11921         18.378         1.5010         0.25367         17.957           33049         11.583         28.413         1.5000         5.0973         1.2124         4.2058         1.7219         18.439         1.0010         0.445697         17.957           33040         11.585         27.464         1.0012         4.4180         18.314         0.8001         0.2567         1.7307         1.5103         18.315         0.8001         0.2567         1.7378         18.913         1.0010         4.2178         2.2070         3.7077         1.5103         18.217         0.6001         0.2178         1.738         1.8121         0.6001         0.2178         1.738           30.010         10.528         2.6473         1.0102         4.4181         1.9381         1.2001         0.7310         1.8212         0.6001         0.11717         1.748           21.001         5.218         5.2018         5.2018         1.8020         0.40003         1.7491         1.7457	p	ρ	$\eta$	p	ρ	$\eta$	p	ρ	$\eta$	р	ρ	η
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	MPa	$mol \cdot dm^{-3}$	µPa•s	MPa	$mol \cdot dm^{-3}$	µPa•s	MPa	$mol \cdot dm^{-3}$	µPa•s	MPa	$mol \cdot dm^{-3}$	µPa•s
53027         11.844         28.931         17.010         6.6234         22.114         4.7499         18.231         18.313         1.001         0.48509         17.944           33.304         11.830         28.518         1.001         0.48509         1.749         4.2986         1.7219         18.313         1.001         0.48509         1.744           33.007         10.552         27.346         1.5004         5.5007         2.7149         1.8135         1.0001         0.48509         1.7597           30.010         10.598         2.6743         1.1051         4.4077         1.5737         1.5138         18.144         0.50012         0.52524         1.7580           30.010         0.8282         2.5811         1.0051         4.4077         1.073         3.2081         1.1448         1.8244         0.50012         0.13637         1.7445           25.014         9.2099         2.5300         8.5009         3.4281         1.9471         2.2001         1.8140         0.40015         0.18170         1.7435           25.014         9.2094         2.8305         1.9411         2.5001         0.6006         1.7570         0.5016         1.1020         0.14130         1.7535						T = 2	98.15 K					
33349         11.880         28.15         16.001         6.2681         21.750         4.4998         1.8229         18.835         1.0012         0.44468         17.355           31.007         10.855         27.346         13.004         1.354         1.012         0.44468         17.355           31.007         10.856         27.346         13.004         1.354         2.6071         1.5193         18.351         0.00112         0.33232         17.856           29.013         10.334         2.6179         1.002         4.4875         2.0079         1.2124         18.141         0.00112         0.32237         17.871           29.013         10.334         2.6179         1.0014         4.4875         2.0009         1.0122         18.144         0.045012         0.23237         17.871           20.014         9.0993         2.5211         8.9993         3.6259         1.9641         2.2500         9.1012         18.146         0.045012         0.1211         17.343           21.000         5.7612         2.3184         0.0931         3.2279         18.341         0.016012         0.2423         18.144         0.04003         0.1111         17.343           21.000         5.2373 <td>35.027</td> <td>11.834</td> <td>28.931</td> <td>17.010</td> <td>6.6254</td> <td>22.114</td> <td>4.7499</td> <td>1.9241</td> <td>18.578</td> <td>1.3001</td> <td>0.52560</td> <td>17.957</td>	35.027	11.834	28.931	17.010	6.6254	22.114	4.7499	1.9241	18.578	1.3001	0.52560	17.957
33004 11-333 28.143 15.006 5.0093 21.232 4.2305 1.7219 18.439 1.0002 0.44416 17316 1	33.949	11.580	28.515	16.001	6.2681	21.756	4.4998	1.8229	18.513	1.2001	0.48509	17.947
1100         11.100         21.44         14.101         1.5110         1.0012         1.1116         1.11116         1.11116         1.11116<	33.004	11.353	28.143	15.006	5.9093	21.424	4.2505	1.7219	18.459	1.1002	0.44468	17.935
50/10         10.598         25/07         1.4160         18.514         0.80/014         0.23232         17.867           28.009         10.062         26.599         1.0005         4.075         20.179         25.030         1.3164         18.254         0.0001         0.23273         17.877           28.009         10.062         25.985         3.044         1.8124         0.0001         0.23273         17.847           26.005         9.9483         25.411         8.9993         1.6250         19.011         2.5003         1.1144         0.35050         1.1147         1.7343           23.008         8.6044         2.2937         7.5016         3.0308         19.212         1.8990         0.76667         18.044         0.30006         0.12111         17.334           23.008         8.6044         2.2937         7.5012         2.218         18.001         0.72811         18.002         0.2001         1.8142         0.30006         1.1161         17.934           21.000         7.3718         2.2848         5.0002         2.2275         18.171         1.5001         0.68763         18.015         0.20001         1.1161         19.343           21.000         7.3718         2.0845 <td>31.995</td> <td>11.105</td> <td>27.743</td> <td>14.004</td> <td>5.5416</td> <td>21.097</td> <td>4.0010</td> <td>1.6208</td> <td>18.409</td> <td>1.0002</td> <td>0.40416</td> <td>17.916</td>	31.995	11.105	27.743	14.004	5.5416	21.097	4.0010	1.6208	18.409	1.0002	0.40416	17.916
$ \begin{array}{c} 22013 \\ 22014 \\ 22005 \\ 2005 \\ 200$	30.010	10.598	26.946	12.002	4.7898	20.790	3.5007	1.4180	18.333	0.80012	0.32324	17.886
	29.013	10.334	26.573	11.005	4.4075	20.179	3.2503	1.3164	18.254	0.70010	0.28277	17.871
$ \begin{array}{c} 27010 & 9.7842 & 25.805 & 9.4094 & 3.817 & 19.735 & 2.7541 & 1.1156 & 18.184 & 0.50012 & 0.20193 & 17.849 \\ 2.5004 & 9.2082 & 25.018 & 8.5009 & 3.4281 & 19.471 & 2.2500 & 0.91063 & 18.096 & 0.45005 & 0.14189 & 17.849 \\ 2.5014 & 2.2091 & 25.0018 & 8.5009 & 3.4281 & 19.471 & 2.2500 & 0.91063 & 18.096 & 0.45005 & 0.1211 & 17.823 \\ 2.2008 & 8.6044 & 24.203 & 7.5016 & 3.0086 & 19.212 & 1.8406 & 0.45005 & 0.2001 & 0.0090 & 17.841 \\ 2.2008 & 8.6044 & 24.203 & 7.5016 & 3.0087 & 1.8001 & 0.75871 & 18.002 & 0.20001 & 0.08071 & 17.840 \\ 2.2000 & 7.6452 & 25.184 & 6.0054 & 2.4309 & 18.844 & 1.5999 & 0.64705 & 18.002 & 0.1000 & 0.04044 & 17.778 \\ 18.008 & 6.7720 & 22.468 & 5.0009 & 2.2271 & 18.711 & 1.001 & 0.60660 & 17.738 & 0.1000 & 0.04044 & 17.778 \\ 35.066 & 0.892 & 2.8535 & 22.011 & 7.5516 & 2.32315 & K \\ 7.6452 & 25.184 & 1.0009 & 6.6497 & 2.32315 & K \\ 35.076 & 0.1872 & 2.464 & 19.007 & 6.6497 & 2.3239 & 4.5042 & 3.1562 & 2.0003 & 3.5026 & 1.1016 & 19.303 \\ 30.101 & 9.7060 & 2.7744 & 19.007 & 6.6497 & 2.3249 & 8.0612 & 2.1766 & 2.0123 & 1.5007 & 0.5584 & 19.097 \\ 30.114 & 9.7086 & 27.143 & 18.000 & 6.3672 & 2.2198 & 8.013 & 2.9470 & 2.0272 & 2.0007 & 0.5584 & 19.097 \\ 30.104 & 9.758 & 2.6464 & 16.009 & 5.6998 & 2.2346 & 6.5036 & 2.4014 & 19.335 & 0.80028 & 0.3768 & 19.040 \\ 30.103 & 9.9506 & 2.714 & 17.007 & 6.6214 & 2.2616 & 7.0093 & 2.5879 & 10.052 & 1.0006 & 0.37239 & 19.013 \\ 30.101 & 9.706 & 2.774 & 17.007 & 6.6214 & 2.2466 & 7.003 & 2.5879 & 1.0052 & 1.0006 & 0.37238 & 19.007 \\ 30.101 & 8.777 & 2.5106 & 5.3710 & 2.2066 & 6.0363 & 2.2178 & 19.806 & 0.6001 & 0.17345 & 19.097 \\ 30.101 & 8.777 & 2.5106 & 5.3710 & 2.2166 & 7.0032 & 2.5879 & 1.0006 & 0.07326 & 18.904 \\ 30.102 & 7.4849 & 2.4510 & 10.007 & 3.6612 & 2.0739 & 0.0002 & 2.5809 & 1.1000 & 0.07326 & 18.904 \\ 30.103 & 9.677 & 2.332 & 2.0104 & 6.6719 & 2.1478 & 4.9004 & 1.4699 & 19.570 & 0.2006 & 0.6774 & 1.8092 \\ 30.101 & 7.4849 & 2.510 & 10.007 & 3.6612 & 2.0739 & 2.0106 & 0.3017 & 0.1007 & 0.13748 & 2.0107 \\ 33.010 & 8.777 & 2.332 & 2.0104 & 6.6719 $	28.009	10.062	26.199	10.001	4.0181	19.882	3.0009	1.2152	18.217	0.60012	0.24235	17.857
$ \begin{array}{c} 2.000 \\ 2.0000 \\ 2.0010 \\ 2.0010 \\ 2.0010 \\ 2.0010 \\ 2.0000 \\ 2.0010 \\ 2.000$	27.010	9.7842	25.805	9.4994	3.8217	19.735	2.7504	1.1136	18.184	0.50012	0.20193	17.849
$ \begin{array}{c} 2.003 \\ 2.004 \\ 2.004 \\ 2.006 \\ 2.004 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.006 \\ 2.007 \\ 2.006 \\ 2.007 \\ 2.006 \\ 2.007 \\ 2.008 \\ 2.006 \\ 2.007 \\ 2.008 \\ 2.000 \\ 2.007 \\ 2.008 \\ 2.000 $	26.005	9.4983	25.411	8.9993	3.6250	19.611	2.5003	1.0122	18.146	0.45005	0.18170	17.843
$ \begin{array}{c} 22008 \\ 8.604 \\ 22001 \\ 8.299 \\ 23013 \\ 7.901 \\ 23.335 \\ 7.002 \\ 7.9701 \\ 23.335 \\ 7.002 \\ 7.9701 \\ 7.318 \\ 22.837 \\ 7.002 \\ 22.835 \\ 7.002 \\ 7.9701 \\ 7.318 \\ 7.002 \\ 22.835 \\ 7.000 \\ 7.9701 \\ 7.318 \\ 7.002 \\ 7.318 \\ 7.000 \\ 7.318 \\ 7.000 \\ 7.318 \\ 7.000 \\ 7.318 \\ 7.000 \\ 7.318 \\ 7.318 \\ 7.000 \\ 7.318$	25.014	9.2099 8.9082	25.030 24.658	8.5009	3.4281	19.471	2.2500	0.91063	18.098	0.40003	0.16149	17.839
22:000         8:2899         23:013         7:0025         2:8313         19:087         1:8:001         0:72819         18:032         0:22001         0:10090         1:7:18:15           21:000         7:6452         23:184         6:0564         2:4:399         18:8:44         1:5999         0:6:6763         18:002         0:10990         0:0001521         17:7:96           18:008         6:3720         22:4:68         5:0005         2:2275         18:7:11         1:5001         0:6:660         17:797         0:10000         0:001841         1:7:778           18:008         6:3720         22:4:68         5:0005         2:275         3:8:711         1:5001         0:5662         0:3003         3:0008         1:156         1:321           31:007         0:424         2:7813         19:990         6:5511         2:333         8:5042         3:126         0:0073         2:057         0:3301         2:156         3:0332         2:5070         0:33012         1:233           31:018         0:438         1:2064         4:3062         2:2161         5:0037         2:0381         1:756         0:41013         0:1483         1:8941           31:014         9:0498         2:1563         5:0379         <	23.008	8.6044	24.293	7.5016	3.0308	19.212	1.8999	0.76867	18.045	0.30005	0.12111	17.824
$ \begin{array}{c} 21000 & 7.9701 & 23.535 & 6.5004 & 2.6299 & 18.968 & 1.7000 & 0.68763 & 18.015 & 0.2000 & 0.080715 & 17.806 \\ 20005 & 7.4518 & 22.847 & 5.5009 & 2.2275 & 18.719 & 1.5001 & 0.66660 & 17.978 & 0.1000 & 0.04034 & 17.778 \\ 18.008 & 6.972 & 22.668 & 5.0005 & 2.2275 & 18.719 & 1.5001 & 0.66660 & 17.978 & 0.1000 & 0.04034 & 17.778 \\ 18.008 & 6.972 & 22.685 & 5.0005 & 2.2244 & 18.608 & 1.4001 & 0.56612 & 17.969 \\ \hline \\ $	22.001	8.2899	23.913	7.0025	2.8313	19.087	1.8001	0.72819	18.032	0.25001	0.10090	17.815
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	21.000	7.9701	23.535	6.5004	2.6299	18.968	1.7000	0.68763	18.015	0.20001	0.080715	17.806
1900b         1.3118         12.837         3.3009         2.2273         18.019         1.3001         0.056012         17.978         0.10000         0.043348         17.78           35.066         10.892         28.525         22.011         7.5514         24.172         9.5004         3.4821         20.502         3.5026         1.3154         1.3303         10.424         27.813         1.999         6.0531         23.338         8.5042         2.3000         0.2414         12.8155         1.9315         1.9315         1.999         0.0311         23.353         8.5042         2.3480         2.0000         0.74443         19.1235           31.997         10.1876         2.7464         19.090         6.6442         2.2328         8.0031         2.3476         2.0070         0.74443         19.163           20.04         9.4558         2.6464         16.099         5.6798         2.2314         6.0336         2.2041         19.935         0.0001         0.74443         18.945           21.004         8.51505         5.3710         2.2050         6.0336         2.2041         19.946         0.30000         0.074461         18.944           25.005         8.392         2.5113         12.006 <td< td=""><td>20.005</td><td>7.6452</td><td>23.184</td><td>6.0054</td><td>2.4309</td><td>18.844</td><td>1.5999</td><td>0.64705</td><td>18.002</td><td>0.14999</td><td>0.060521</td><td>17.794</td></td<>	20.005	7.6452	23.184	6.0054	2.4309	18.844	1.5999	0.64705	18.002	0.14999	0.060521	17.794
$ \begin{array}{c} 13003 & 1.4001 & 0.37236 & 1.8001 & 1.4001 & 0.37236 & 1.8001 & 1.4001 & 0.37236 & 1.8001 & 1.4001 & 0.37236 & 1.8001 & 1.4001 & 1.4001 & 1.4001 & 1.4001 & 0.37236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0014 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.037236 & 1.0024 & 0.0018 & 0.0014 & 0.037236 & 1.0024 & 0.0018 & 0.0014 & 0.037236 & 1.0024 & 0.0018 & 0.0014 & 0.037236 & 1.0024 & 0.0018 & 0.0014 & 0.037236 & 1.0024 & 0.0018 & 0.0014 & 0.037236 & 1.0024 & 0.0018 & 0.0014 & 0.037236 & 1.0024 & 0.0018 & 0.0014 & 0.037236 & 1.0024 & 0.0018 & 0.0024 & 0.0018 & 0.0024 & 0.0018 & 0.0024 & 0.0018 & 0.0014 & 0.0037236 & 0.0018 & 0.0018 & 0.0018 & 0.001$	19.006	7.3118	22.837	5.5009	2.2275	18.719	1.5001	0.60660	17.978	0.10000	0.040348	17.778"
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	10.000	0.9720	22.400	5.0005	2.0234	18.008	1.4001	0.30012	17.909			
3.000         10.82         28.252         22.010         7.3543         24.123         2.3004         2.3004         1.3006         1.3006 <td>25.000</td> <td>10.002</td> <td>20 525</td> <td>22.011</td> <td>7 5514</td> <td>T = 3</td> <td>23.15 K</td> <td>2 4921</td> <td>20 (20</td> <td>2 5026</td> <td>1 2014</td> <td>10.400</td>	25.000	10.002	20 525	22.011	7 5514	T = 3	23.15 K	2 4921	20 (20	2 5026	1 2014	10.400
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	32.000	10.892	20.525 28.135	22.011	7 2565	24.172	9.5004 9.0061	3.4821 3.3063	20.620	3.0020	1.5014	19.400
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	33.007	10.424	27.813	19,999	6.9531	23.538	8.5042	3.1269	20.300	2.5009	0.93012	19.221
31.013       9.9506       27,143       18.000       6.3362       22.918       7.5027       2.7665       20.153       1.5007       0.558455       19.031         29.014       9.4558       26.464       16.009       5.6998       22.334       6.5036       2.4041       19.935       0.80028       0.29786       19.031         29.014       9.4558       26.464       16.009       5.0783       2.1763       5.5039       2.0388       19.756       0.40011       0.22336       18.983         27.000       8.9361       25.785       14.005       5.0383       21.763       5.5039       2.0388       19.756       0.40013       0.14893       18.961         25.005       8.3992       25.113       12.006       4.3586       21.243       4.5004       1.6699       19.577       0.10004       0.037236       18.904         23.013       7.8409       24.510       10.007       3.6612       20.739       2.332       2.1033       3.5042       2.1336       2.0236       2.0336         23.013       9.4251       27.466       19.006       6.3944       2.4037       7.4183       2.0162       2.332       2.2103       0.3042       2.0327       2.0232       2.0320 <td< td=""><td>31.997</td><td>10.187</td><td>27.464</td><td>19.007</td><td>6.6497</td><td>23.229</td><td>8.0031</td><td>2.9470</td><td>20.272</td><td>2.0007</td><td>0.74436</td><td>19.168</td></td<>	31.997	10.187	27.464	19.007	6.6497	23.229	8.0031	2.9470	20.272	2.0007	0.74436	19.168
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	31.013	9.9506	27.143	18.000	6.3362	22.918	7.5027	2.7665	20.153	1.5007	0.55845	19.097
29.014 9.4558 26.464 16.009 5.6998 22.334 6.5036 2.2404 19.935 0.80028 0.29786 19.004 28.012 9.1999 26.128 15.005 5.3710 22.055 6.0363 2.2217 19.826 0.60011 0.2233 18.961 25.005 8.5992 25.113 12.006 4.3866 21.243 4.5002 1.8546 19.648 0.9009 0.11169 18.946 25.005 8.5992 25.113 12.006 4.3866 21.243 4.5002 1.8546 19.648 0.9009 0.074461 18.934 23.013 7.8409 24.510 10.007 3.6615 20.730 $T=338.15  {\rm K}$ 34.919 10.063 28.332 22.011 6.9470 24.577 9.4989 3.2036 21.503 3.5042 1.2036 20.439 34.013 9.8677 28.075 21.004 6.6718 24.037 8.5018 2.2170 0.1004 0.037236 18.904 33.000 9.6457 27.762 20.006 6.3944 24.037 8.5018 2.8780 21.292 2.5020 0.86112 20.298 32.013 9.4251 27.466 19.008 6.1121 23.781 8.0012 2.7177 21.196 2.0016 0.6853 20.228 31.015 9.1981 2.7170 18.004 5.8336 23.508 7.5025 2.5486 21.009 1.5019 0.51782 20.167 33.000 8.7672 2.6546 16.001 5.2343 22.993 6.5013 2.2156 20.108 1.0009 0.43452 20.167 30.001 8.9675 2.6872 17.002 5.5313 22.575 7.002 2.3826 21.009 1.5019 0.51782 20.167 37.005 8.2414 25.983 14.001 4.6281 22.498 5.5028 1.8808 20.742 0.40019 0.4358 20.114 29.000 8.7262 26.546 16.001 5.2343 22.993 6.5013 2.2156 20.198 0.80057 0.27629 20.073 77.053 8.2414 25.983 14.001 4.6281 22.498 5.5028 1.8808 20.742 0.40019 0.1318 20.051 25.007 7.7373 25.431 12.007 4.0070 22.034 4.5082 1.5449 20.883 0.20008 0.069102 20.029 23.006 7.2140 24.864 10.001 3.3661 21.608  T =373.15 K 34.834 9.3680 28.444 22.011 5.61892 24.868 9.0021 2.5118 2.2012 0.46277 21.033 3.0035 0.96111 21.372 3.0003 8.3366 2.7138 17.018 5.1293 23.920 7.0069 2.2133 21.944 1.0005 0.32577 21.402 43.864 10.001 3.3661 21.608 3.072 2.1073 2.0519 0.3005 0.5755 21.120 3.0013 8.3706 2.0005 23.006 7.2140 24.864 10.001 3.3661 21.608 3.072 2.0156 3.00013 0.103456 20.004 3.001 8.772 2.7653 19.005 5.6641 24.400 8.0027 2.5182 22.102 3.0013 0.103456 20.004 3.001 8.5396 2.4444 2.2011 5.6433 2.5097 2.1270 3.0055 0.25579 2.1.203 3.003 8.3366 2.7138 17.018 5.1293 23.920 7.0069 2.2133 21.944 1.0005 0.32177 21.138 3.000 8.8258 28.742 2.0019 5.7584 23.404 8.0027 2.5182 22.102 3.0	30.014	9.7060	26.774	17.007	6.0214	22.616	7.0093	2.5879	20.052	1.0006	0.37239	19.031
$ \begin{array}{c} 28.012 & 9.1999 & 26.128 & 15.005 & 5.3710 & 22.050 & 6.0366 & 2.2217 & 19.286 & 0.60011 & 0.22336 & 18.983 \\ 27.000 & 8.0361 & 25.887 & 14.005 & 5.0383 & 21.763 & 5.5039 & 2.0388 & 19.576 & 0.40013 & 0.14893 & 18.966 \\ 26.011 & 8.6726 & 25.472 & 13.005 & 4.7006 & 21.510 & 5.0022 & 1.8846 & 19.464 & 0.30009 & 0.11169 & 18.946 \\ 25.005 & 8.2992 & 25.113 & 12.006 & 4.3386 & 21.243 & 4.5004 & 1.6699 & 19.570 & 0.2006 & 0.0774461 & 18.934 \\ 24.004 & 8.1214 & 24.806 & 11.007 & 4.0124 & 20.977 & 4.0118 & 1.4860 & 19.477 & 0.10004 & 0.037236 & 18.904 \\ 32.013 & 7.8409 & 24.510 & 10.007 & 3.6615 & 20.730 & $$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$	29.014	9.4558	26.464	16.009	5.6998	22.334	6.5036	2.4041	19.935	0.80028	0.29786	19.004
$ \begin{array}{c} 2000 & 8.201 & 25.75 & 14.005 & 5.003 & 21.60 & 5.002 & 1.8546 & 19.648 & 0.4003 & 0.14893 & 18.946 \\ 25.005 & 8.3992 & 25.113 & 12.006 & 4.3586 & 21.510 & 5.0022 & 1.8546 & 19.648 & 0.3009 & 0.074461 & 18.944 \\ 25.005 & 8.3992 & 25.113 & 12.006 & 4.3586 & 21.43 & 4.3004 & 1.6699 & 19.477 & 0.1004 & 0.073236 & 18.904 \\ 23.013 & 7.8409 & 24.510 & 10.007 & 3.6615 & 20.730 & $$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$$	28.012	9.1999	26.128	15.005	5.3710	22.050	6.0036	2.2217	19.826	0.60011	0.22336	18.983
$ \begin{array}{c} 2.016 \\ 2.5004 \\ 8.1992 \\ 2.5113 \\ 2.006 \\ 1.2100 \\ 1.007 \\ 1.0004 \\ 1.007 \\ 1.0004 \\ 1.007 \\ 1.0004 \\ 1.007 \\ 1.0004 \\ 1.007 \\ 1.0004 \\ 1.007 \\ 1.0004 \\ 1.0004 \\ 1.007 \\ 1.0004 \\ 1.0004 \\ 1.007 \\ 1.0004 \\ 1.0004 \\ 1.007 \\ 1.0004$	27.000	8.9301	25.785	14.005	5.0385	21.703	5.0039	2.0388	19.750	0.40013	0.14895	18.901
$ \begin{array}{c} 24,004 \\ 8,1214 \\ 24,905 \\ 11,007 \\ 24,510 \\ 10,007 \\ 3,6615 \\ 22,013 \\ 7,8409 \\ 24,510 \\ 10,007 \\ 24,515 \\ 10,007 \\ 24,515 \\ 24,016 \\ 24,577 \\ 24,577 \\ 24,515 \\ 24,016 \\ 24,577 \\ 24,578 \\ 24,578 \\ 24,577 \\ 24,578 \\ 24,5$	25.005	8 3992	25.472	12 006	4.7000	21.510	4 5004	1.6540	19.048	0.30009	0.074461	18 934
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	24.004	8.1214	24.806	11.007	4.0124	20.977	4.0018	1.4860	19.477	0.10004	0.037236	$18.904^{a}$
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	23.013	7.8409	24.510	10.007	3.6615	20.730						
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						T = 3	48 15 K					
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	34.919	10.063	28.332	22.011	6.9470	24.577	9.4989	3.2036	21.503	3.5042	1.2036	20.439
33.000 9.6457 27.762 20.006 6.3944 24.037 8.5018 2.8780 21.292 2.5020 0.86112 20.298 33.013 9.4251 27.466 19.008 6.1121 23.781 8.0021 2.7137 21.196 2.0016 0.68953 20.228 31.015 9.1981 27.170 18.004 5.8236 23.508 7.5025 2.5486 21.099 1.5019 0.51782 20.167 30.021 8.9675 26.872 17.002 5.5313 23.255 7.0023 2.3826 21.008 1.0009 0.34535 20.111 29.00 8.7262 26.546 16.001 5.2343 22.993 6.5013 2.2156 20.918 0.80057 0.27629 20.087 27.995 8.4841 26.283 15.011 4.9366 22.742 6.0034 2.0489 20.836 0.60046 0.20729 20.073 27.005 8.2414 25.983 14.001 4.6281 22.498 5.5022 1.7120 20.659 0.30013 0.10365 20.041 25.007 7.7373 25.431 12.007 4.0070 22.034 4.5082 1.5449 20.836 0.6004 0.20729 20.005 23.006 7.2140 24.864 10.001 3.3661 21.608 T = 373.15 K 34.834 9.3680 28.444 22.011 6.64433 25.098 9.4991 2.9710 22.383 3.5033 1.1195 21.436 33.010 8.5591 77.411 8.001 5.3960 24.468 9.0021 2.8214 22.279 3.0035 0.96111 21.372 33.011 8.9900 27.920 19.977 5.9200 24.617 8.5043 2.6707 2.2199 2.5014 0.80154 21.310 33.010 8.5591 77.411 18.001 5.3960 24.468 9.0021 2.8214 22.279 3.0035 0.96111 21.372 33.010 8.5591 7.7411 18.001 5.3960 24.424 7.5007 2.5182 22.112 2.0024 0.64237 21.253 31.001 8.5591 7.7411 18.001 5.3960 23.700 7.25182 22.112 2.002 0.64237 21.253 31.001 8.5591 7.7411 8.0001 5.3960 23.700 7.25182 22.1877 0.80056 0.25759 21.120 28.007 7.8916 26.616 15.013 4.8530 23.701 6.5074 2.0592 21.877 0.80056 0.25759 21.120 28.007 7.8916 26.616 15.013 4.8530 23.701 6.5074 2.0592 21.877 0.80056 0.25759 21.120 28.007 7.8916 26.616 15.013 4.8530 23.701 6.5074 2.0592 21.877 0.80056 0.25759 21.120 28.017 7.8916 26.616 15.013 4.8530 23.701 6.5074 2.0592 21.877 0.80056 0.25759 21.120 28.007 7.881 26.345 14.010 4.2908 23.270 5.5047 1.5927 21.636 0.30098 0.096950 21.080 25.012 7.148 25.838 12.000 3.7114 22.845 4.5016 1.4341 21.570 0.20008 0.064462 21.061 24.005 6.3999 25.586 11.006 3.4195 22.670 4.0100 1.2795 21.503 0.10025 0.032307 21.025 33.034 8.4292 28.221 20.019 5.5352 25.246 8.5017 4.1591 23.244 3.5018 1.0466 22.418 34.043 8.6301 28.425 21.001 5.7352 25.2	34.013	9.8677	28.075	21.004	6.6718	24.308	9.0040	3.0424	21.392	3.0026	1.0324	20.367
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	33.000	9.6457	27.762	20.006	6.3944	24.037	8.5018	2.8780	21.292	2.5020	0.86112	20.298
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	32.013	9.4251	27.466	19.008	6.1121	23.781	8.0021	2.7137	21.196	2.0016	0.68953	20.228
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	31.015	9.1981	27.170	18.004	5.8236	23.508	7.5025	2.5486	21.099	1.5019	0.51782	20.167
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	30.021	8.9675	26.872	17.002	5.5313	23.255	6 5012	2.3826	21.008	1.0009	0.34535	20.111
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	29.000	8.7202	26.340	15 011	2.2343 4.9366	22.993	6.0034	2.2130	20.916	0.80037	0.27029	20.087
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	27.005	8.2414	25.983	14.001	4.6281	22.498	5.5028	1.8808	20.742	0.40019	0.13818	20.073
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	26.007	7.9919	25.709	13.006	4.3201	22.267	5.0022	1.7120	20.659	0.30013	0.10365	20.041
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	25.007	7.7373	25.431	12.007	4.0070	22.034	4.5082	1.5449	20.583	0.20008	0.069102	20.029
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	24.004	7.4775	25.145	11.007	3.6893	21.817	3.9996	1.3722	20.509	0.10007	0.034566	$20.005^{a}$
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	23.006	7.2140	24.864	10.001	3.3661	21.608						
$\begin{array}{cccccccccccccccccccccccccccccccccccc$						$T = 3^{\circ}$	73.15 K					
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	34.834	9.3680	28.444	22.011	6.4433	25.098	9.4991	2.9710	22.383	3.5033	1.1195	21.436
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	54.006 33.011	9.1977	28.207	21.015	6.1892	24.868	9.0021	2.8214	22.279	3.0035	0.96111	21.372
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	32 010	0.9900 8 7770	27.920	19.977	5.9200 5.66/1	24.017 24.400	0.3043 8 0027	2.0707	22.199 22.199	2.3014	0.60134	21.510 21.253
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	31.001	8.5591	27.411	18.001	5.3960	24.142	7.5007	2.3648	22.026	1.5012	0.48227	21.203
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	30.003	8.3396	27.138	17.018	5.1293	23.920	7.0069	2.2133	21.944	1.0005	0.32177	21.138
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	29.005	8.1164	26.884	16.013	4.8530	23.701	6.5074	2.0592	21.877	0.80056	0.25759	21.120
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	28.017	7.8916	26.616	15.013	4.5742	23.480	6.0006	1.9023	21.797	0.60066	0.19336	21.104
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	27.007	7.6581	26.345	14.010	4.2908	23.270	5.5047	1.7480	21.718	0.40017	0.12887	21.087
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	26.006	7.4225	26.099	13.002	4.0020	23.068	5.0074	1.5927	21.636	0.30098	0.096950	21.080
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	25.012	/.1848	23.838 25.594	12.000	5./114 3./105	22.845	4.5016	1.4341	21.570	0.20008	0.004462	21.001 21.025a
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	24.003	6 6925	25.360	10.010	3 1238	22.070	4.0100	1.2795	21.303	0.10025	0.032307	21.025
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	20.004	0.0725	20.004	10.010	5.1250	T A	00 15 17					
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	35.040	8 8758	28 724	22.011	6.0157	I = 3	98.13 K 9 5060	2 7748	23 244	3 5018	1 0466	22 /18
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	34.043	8.6301	28.452	21.009	5.7758	25.461	9.0103	2.6357	23.173	3.0007	0.89832	22.359
32.013       8.2225       27.956       19.003       5.2846       25.033       8.0076       2.3524       23.020       2.0013       0.60103       22.252         31.023       8.0190       27.729       18.010       5.0365       24.822       7.5071       2.2100       22.944       1.5018       0.45171       22.204         29.911       7.7866       27.472       17.010       4.7831       24.631       7.0053       2.0665       22.871       1.0010       0.30153       22.153         29.028       7.5992       27.279       16.005       4.5250       24.434       6.5018       1.9218       22.796       0.80046       0.24126       22.131         28.028       7.3836       27.028       15.013       4.2665       24.225       6.0049       1.7784       22.734       0.60027       0.18103       22.102         25.977       6.9313       26.566       13.002       3.7332       23.864       5.0037       1.4875       22.601       0.30079       0.090788       22.087         25.001       6.7111       26.336       11.989       3.4594       23.685       4.5031       1.3411       22.537       0.20088       0.060649       22.072         23.993 <td< td=""><td>33.034</td><td>8.4292</td><td>28.221</td><td>20.019</td><td>5.5352</td><td>25.246</td><td>8.5010</td><td>2.4922</td><td>23.106</td><td>2.4958</td><td>0.74839</td><td>22.305</td></td<>	33.034	8.4292	28.221	20.019	5.5352	25.246	8.5010	2.4922	23.106	2.4958	0.74839	22.305
31.023       8.0190       27.729       18.010       5.0365       24.822       7.5071       2.2100       22.944       1.5018       0.45171       22.204         29.911       7.7866       27.472       17.010       4.7831       24.631       7.0053       2.0665       22.871       1.0010       0.30153       22.153         29.028       7.5992       27.279       16.005       4.5250       24.434       6.5018       1.9218       22.796       0.80046       0.24126       22.131         28.028       7.3836       27.028       15.013       4.2665       24.225       6.0049       1.7784       22.734       0.60027       0.18103       22.102         25.977       6.9313       26.566       13.002       3.7332       23.864       5.0037       1.4875       22.601       0.30079       0.090788       22.087         25.001       6.7111       26.336       11.989       3.4594       23.685       4.5031       1.3411       22.537       0.20088       0.060649       22.072         23.993       6.4801       26.102       11.003       3.1897       23.502       4.0028       1.1942       22.474       0.10008       0.030225       22.033         23.034       <	32.013	8.2225	27.956	19.003	5.2846	25.033	8.0076	2.3524	23.020	2.0013	0.60103	22.252
29.911       7.7866       27.472       17.010       4.7831       24.631       7.0053       2.0665       22.871       1.0010       0.30153       22.153         29.028       7.5992       27.279       16.005       4.5250       24.434       6.5018       1.9218       22.796       0.80046       0.24126       22.131         28.028       7.3836       27.028       15.013       4.2665       24.225       6.0049       1.7784       22.734       0.60027       0.18103       22.124         27.031       7.1655       26.793       13.997       3.9988       24.047       5.5026       1.6327       22.661       0.40045       0.12083       22.102         25.977       6.9313       26.566       13.002       3.7332       23.864       5.0037       1.4875       22.601       0.30079       0.090788       22.087         25.001       6.7111       26.336       11.989       3.4594       23.685       4.5031       1.3411       22.537       0.20088       0.060649       22.072         23.993       6.4801       26.102       11.003       3.1897       23.502       4.0028       1.1942       22.474       0.10008       0.030225       22.033         23.034	31.023	8.0190	27.729	18.010	5.0365	24.822	7.5071	2.2100	22.944	1.5018	0.45171	22.204
29.028       7.5992       27.279       16.005       4.5250       24.434       6.5018       1.9218       22.796       0.80046       0.24126       22.131         28.028       7.3836       27.028       15.013       4.2665       24.225       6.0049       1.7784       22.734       0.60027       0.18103       22.124         27.031       7.1655       26.793       13.997       3.9988       24.047       5.5026       1.6327       22.661       0.40045       0.12083       22.102         25.977       6.9313       26.566       13.002       3.7332       23.864       5.0037       1.4875       22.601       0.30079       0.090788       22.087         25.001       6.7111       26.336       11.989       3.4594       23.685       4.5031       1.3411       22.537       0.20088       0.060649       22.072         23.993       6.4801       26.102       11.003       3.1897       23.502       4.0028       1.1942       22.474       0.10008       0.030225       22.033         23.034       6.2570       25.896       10.005       2.9137       23.337       1.942       22.474       0.10008       0.030225       22.033	29.911	7.7866	27.472	17.010	4.7831	24.631	7.0053	2.0665	22.871	1.0010	0.30153	22.153
28.028         7.3836         27.028         15.013         4.2665         24.225         6.0049         1.7784         22.734         0.60027         0.18103         22.124           27.031         7.1655         26.793         13.997         3.9988         24.047         5.5026         1.6327         22.661         0.40045         0.12083         22.102           25.977         6.9313         26.566         13.002         3.7332         23.864         5.0037         1.4875         22.601         0.30079         0.090788         22.087           25.001         6.7111         26.336         11.989         3.4594         23.685         4.5031         1.3411         22.537         0.20088         0.060649         22.072           23.993         6.4801         26.102         11.003         3.1897         23.502         4.0028         1.1942         22.474         0.10008         0.030225         22.033           23.034         6.2570         25.896         10.005         2.9137         23.337         23.337	29.028	7.5992	27.279	16.005	4.5250	24.434	6.5018	1.9218	22.796	0.80046	0.24126	22.131
27.051       7.1053       26.795       13.997       5.9988       24.047       5.5026       1.6527       22.601       0.40045       0.12083       22.102         25.977       6.9313       26.566       13.002       3.7332       23.864       5.0037       1.4875       22.601       0.30079       0.090788       22.087         25.001       6.7111       26.336       11.989       3.4594       23.685       4.5031       1.3411       22.537       0.20088       0.060649       22.072         23.993       6.4801       26.102       11.003       3.1897       23.502       4.0028       1.1942       22.474       0.10008       0.030225       22.033         23.034       6.2570       25.896       10.005       2.9137       23.337	28.028	7.3836	27.028	15.013	4.2665	24.225	6.0049	1.7784	22.734	0.60027	0.18103	22.124
22.577         0.5213         20.500         15.002         5.7522         25.004         5.0057         1.4875         22.001         0.50079         0.990788         22.087           25.001         6.7111         26.336         11.989         3.4594         23.685         4.5031         1.3411         22.537         0.20088         0.060649         22.072           23.993         6.4801         26.102         11.003         3.1897         23.502         4.0028         1.1942         22.474         0.10008         0.030225         22.033           23.034         6.2570         25.896         10.005         2.9137         23.337         23.337	21.031	/.1055	20.193 26.566	13.997	3.9988 3.7320	24.047	5.5026	1.032/ 1.4275	22.001	0.40045	0.12083	22.102
23.993 6.4801 26.102 11.003 3.1897 23.502 4.0028 1.1942 22.474 0.10008 0.030225 22.033 23.034 6.2570 25.896 10.005 2.9137 23.337	25.977	67111	26.300	11 989	3.7332 3.4594	23.604	4 5031	1.4073	22.001	0.20079	0.050788	22.087
23.034 6.2570 25.896 10.005 2.9137 23.337	23.993	6.4801	26.102	11.003	3.1897	23.502	4.0028	1.1942	22.337	0.10008	0.030225	22.033ª
	23.034	6.2570	25.896	10.005	2.9137	23.337						

Table 1 (Continued)

р	ρ	η	р	ρ	η	р	ρ	η	р	ρ	η
MPa	mol·dm <sup>-3</sup>	µPa•s	MPa	mol·dm <sup>-3</sup>	µPa•s	MPa	mol·dm <sup>-3</sup>	µPa•s	MPa	mol·dm <sup>-3</sup>	µPa•s
					T = 4	23.15 K					
35.050	8.3185	29.033	22.009	5.6469	26.302	9.5043	2.6026	24.122	3.5058	0.98438	23.367
34.020	8.1254	28.815	20.987	5.4160	26.092	9.0042	2.4712	24.052	3.0023	0.84459	23.322
32.989	7.9290	28.585	20.008	5.1918	25.913	8.5030	2.3389	23.978	2.4980	0.70402	23.271
31.988	7.7356	28.366	19.008	4.9598	25.726	8.0042	2.2066	23.914	1.9978	0.56405	23.223
31.001	7.5421	28.155	17.999	4.7224	25.539	7.4979	2.0716	23.842	1.5025	0.42494	23.177
30.011	7.3450	27.942	17.009	4.4864	25.354	7.0030	1.9390	23.775	1.0011	0.28362	23.132
29.015	7.1441	27.736	16.009	4.2451	25.174	6.5023	1.8041	23.720	0.80032	0.22688	23.117
28.010	6.9384	27.529	15.002	3.9991	25.005	6.0022	1.6688	23.651	0.60023	0.17027	23.104
27.035	6.7360	27.323	14.011	3.7539	24.832	5.5028	1.5331	23.583	0.40013	0.11358	23.089
26.017	6.5215	27.107	13.002	3.5014	24.665	5.0027	1.3966	23.532	0.30044	0.085312	23.069
25.024	6.3095	26.911	11.994	3.2458	24.506	4.5012	1.2591	23.477	0.20007	0.056830	23.054
24.019	6.0917	26.691	11.004	2.9923	24.344	4.0040	1.1221	23.417	0.10005	0.028427	$23.018^{a}$
23.014	5.8707	26.505	10.006	2.7337	24.185						

<sup>a</sup> Influenced by slip.



**Figure 2.** Evaluation of all isotherms with a double polynomial series expansion of the fourth order in the reduced density ( $\delta$ ) and of the second order in the reciprocal reduced temperature ( $\tau$ ) (eq 2). Deviations [ $\Delta = 100(\eta_{exp} - \eta_{cor})/\eta_{cor}$ ] as a function of molar density ( $\rho$ ).  $\bigcirc$ , 298.15 K;  $\triangle$ , 323.15 K;  $\heartsuit$ , 348.15 K;  $\bigtriangledown$ , 373.15 K;  $\square$ , 398.15 K;  $\times$ , 423.15 K.

is a further evidence of the high reproducibility of the present measurements.

### **Comparison with Literature**

Several complete viscosity surface correlations are available for nitrogen (e.g., those by Stephan et al.<sup>2</sup> and by Millat and Vesovic<sup>3</sup> as well as the most recent one by Lemmon and Jacobsen<sup>4</sup>). As already discussed in the Introduction, the correlation of Lemmon and Jacobsen has been chosen as basis for a comparison between the present data and those of the literature.

Figure 3 illustrates for the dilute-gas region the deviations of reliable literature data as well as of the present data from the correlation by Lemmon and Jacobsen. The measurements were performed with five different techniques. Smith and co-workers<sup>15–18</sup> used capillary viscometers for relative measurements, whereas Hoogland et al.<sup>19</sup> applied an absolute one. Kestin et al.,<sup>20–22</sup> Timrot et al.,<sup>23</sup> and Vogel and his collaborators<sup>24,25</sup> used oscillating-disk viscometers in a relative manner. Docter et al.<sup>26,27</sup> and Evers et al.<sup>28</sup> developed a newly designed rotating-cylinder viscometer combined with a densimeter for absolute measurements. Hurly et al.<sup>29</sup> performed their measurements with a relative Greenspan acoustic viscometer. Figure 3 shows that all these data, including those of the present paper, agree with the correlation of Lemmon and Jacobsen at atmospheric or lower



**Figure 3.** Comparison of reliable experimental low-density viscosity data with the correlation of Lemmon and Jacobsen.<sup>4</sup> Deviations [ $\Delta = 100(\eta_{exp} - \eta_{cor})/\eta_{cor}$ ] as a function of temperature (*T*). Experimental data:  $\bigcirc$ , Clarke and Smith;<sup>15</sup>  $\square$ , Dawe and Smith;<sup>16</sup>  $\boxplus$ , Gough et al.;<sup>17</sup>  $\otimes$ , Matthews et al.;<sup>18</sup>  $\bigstar$ , Hoogland et al.;<sup>19</sup> #, Kestin et al.;<sup>20</sup> <, Kestin et al.;<sup>21</sup> >, Kestin et al.;<sup>22</sup> +, Timrot et al.;<sup>23</sup>  $\bigtriangledown$ , Vogel;<sup>24</sup>  $\square$ , Vogel et al.;<sup>25</sup>  $\diamond$ , Docter et al.;<sup>26,27</sup>  $\triangle$ , Evers et al.;<sup>28</sup> ×, Hurly et al.;<sup>29</sup>  $\bigcirc$ , present work.

pressures within  $\pm$  0.4 %. The experimental data of Kestin et al.,<sup>20,21,22</sup> determined at temperatures higher than room temperature, show systematic positive deviations from the other data. This is in agreement with findings for noble gases and is probably due to a temperature measurement error in the experimental equipment of Kestin and co-workers extensively discussed by Vogel et al.<sup>30</sup> Furthermore, it is to consider that Smith and co-workers<sup>15-18</sup> directed their measurements to the ranges of very high and low temperatures connected with uncertainties of about  $\pm$  0.5 % at the temperatures under discussion. Hence, it can be concluded that the most reliable data19,23-25,28 including those of the present paper even agree within  $\pm$  (0.1 to 0.2) % between room temperature and 420 K. The difference between the basis correlation of Lemmon and Jacobsen and the most reliable data is probably caused by that Lemmon and Jacobsen do not critically evaluate the uncertainties of the measured data, which are included in the development of their correlation.

In Figure 4, the experimental data of the present paper are compared with the correlation of Lemmon and Jacobsen<sup>4</sup> for all six isotherms in the complete density range up to a maximum of 11.8 mol·dm<sup>-3</sup>. Systematic deviations between the correlation by Lemmon and Jacobsen and the present data become evident in the limit of zero density as already shown in Figure 3.



**Figure 4.** Comparison of the experimental viscosity data of the present paper at different temperatures with the viscosity surface correlation of Lemmon and Jacobsen.<sup>4</sup> Deviations [ $\Delta = 100(\eta_{exp} - \eta_{cor})/\eta_{cor}$ ] as a function of molar density ( $\rho$ ).  $\bigcirc$ , 298.15 K;  $\triangle$ , 323.15 K;  $\diamondsuit$ , 348.15 K;  $\bigtriangledown$ , 373.15 K;  $\Box$ , 398.15 K;  $\times$ , 423.15 K.



**Figure 5.** Comparison of the experimental viscosity data of the present paper at different temperatures with the viscosity surface correlations of Stephan et al.<sup>2</sup> (open symbols) and of Millat and Vesovic<sup>3</sup> (filled symbols). Deviations [ $\Delta = 100(\eta_{exp} - \eta_{cor})/\eta_{cor}$ ] as a function of molar density ( $\rho$ ).  $\bigcirc$ ,  $\oplus$ , 298.15 K;  $\triangle$ ,  $\blacktriangle$ , 323.15 K;  $\diamondsuit$ ,  $\blacklozenge$ , 348.15 K;  $\bigtriangledown$ ,  $\blacktriangledown$ , 373.15 K;  $\Box$ ,  $\blacksquare$ , 398.15 K;  $\times$ , #, 423.15 K.

Furthermore, Figure 4 reveals at moderate densities a slight difference in the initial density dependence of the viscosity. Taking into account these findings, the density dependence of the new experimental data agrees nearly perfectly with that of the correlation of Lemmon and Jacobsen up to densities of about 8 mol·dm<sup>-3</sup>. But at higher densities, it is obvious that the density dependence of the viscosity differs somewhat systematically. To investigate this in more detail, the new experimental values have also been compared with directly measured data used for the development of the surface correlation of Lemmon and Jacobsen (see below). All in all, the new data show only deviations of  $\pm$  0.7 % from the correlated values in the temperature and density ranges of the measurements.

The comparison of the new experimental data with the other correlations illustrated in Figure 5 makes clear that the representation of the viscosity surface of nitrogen by Stephan et al.<sup>2</sup> essentially corresponds to that by Lemmon and Jacobsen. On the contrary, the correlation of Millat and Vesovic<sup>3</sup> disagrees completely with the other ones. The reason for that cannot be

detected, because the publication of Millat and Vesovic does not provide detailed information about their calculations.

The comparison with the directly measured data from the literature is somewhat complicated since the evaluation of the measurements requires the density of the fluid and consequently an equation of state that was not the same in different papers. In addition, the viscosity data were sometimes reported as a function of temperature and pressure, but not of density. In the case that the densities were given by the authors, we used them immediately. Alternatively, we calculated ourselves the densities applying the equation of state by Span et al.<sup>1</sup> The experimental values of the present paper correlated with the double polynomial in density and temperature discussed in the preceding section are used for the comparison. First, we will discuss data restricted to densities up to about 6 mol·dm<sup>-3</sup>.

Figure 6 shows the deviations from the experimental data of Kestin et al.,14,31,32 all measured with oscillating-disk viscometers. Kestin and Leidenfrost14 performed absolute measurements according to the theory of Newell.33 Here, some data at the highest densities could already be affected by that the validity range of the measuring theory has been exceeded. The relative measurements of Kestin et al.32 may be characterized at higher densities by larger uncertainties too. The reason is that the data of Kestin and Leidenfrost14 were used for calibration and that the calibration curve was extrapolated to higher densities according to an inappropriate theory.34-36 Furthermore, the relative measurements of Kestin and Leidenfrost<sup>31</sup> were based on older data for calibration in a large density range. Considering that our vibrating-wire viscometer has been calibrated with viscosity values of Kestin and Leidenfrost14 in the density range up to 1 mol·dm<sup>-3</sup>, it is to be noticed that the deviations enlarge systematically with increasing densities. Nevertheless, the deviations are generally within  $\pm 0.4$  % up to a density of 6.3 mol·dm<sup>-3</sup> apart from one outlier.

The relative measurements of Timrot et al.<sup>23</sup> at temperatures between (295 and 389) K already discussed for the low-density region were also performed to densities up to nearly 5 mol·dm<sup>-3</sup>. It is remarkable that, despite a slightly inferior reproducibility, the deviations between these data and the new experimental values are within  $\pm 0.4$  % up to a density of 3 mol·dm<sup>-3</sup> as shown in Figure 7. Only two data are characterized by larger differences. In addition, this figure demonstrates that the results of Hoogland et al.,<sup>19</sup> derived from absolute measurements with their capillary viscometer in the temperature range (298 to 333) K at densities up to 5 mol·dm<sup>-3</sup>, do not deviate from the present data by more than  $\pm$  0.4 %. Furthermore, Figure 7 illustrates that the relative measurements with an acoustic Greenspan viscometer by Hurly et al.<sup>29</sup> at room temperature and at moderate densities up to 1.3 mol·dm<sup>-3</sup> resulted in values that are approximately 0.2 % lower than the present data.

Michels and Gibson<sup>37</sup> obtained reliable results with a capillary viscometer considering that these absolute measurements between (298 and 348) K at densities up to 20 mol·dm<sup>-3</sup> were already performed in 1932. The differences between the data of Michels and Gibson and the present values do not exceed  $\pm$  0.4 up to 6 mol·dm<sup>-3</sup> as shown in Figure 8. The deviations increase up to -1.5 % at higher densities up to 12 mol·dm<sup>-3</sup> adherent with a larger scattering of the data of Michels and Gibson.

Figure 9 presents the results of the comparison with the data of Flynn et al.<sup>38</sup> and Gracki et al.,<sup>39</sup> both measured with an absolute capillary viscometer. The values of the present paper agree with the data of Flynn et al. within  $\pm$  0.4 %, which means up to densities of about 6.8 mol·dm<sup>-3</sup>. The data of the 298 K

Table 2. Coefficients of Equation 1

Т		$ ho_{ m max}$	$\eta_0$	$\eta_1$	$\eta_2$	$\eta_3$	$\eta_4$	
K	п	mol·dm <sup>-3</sup>	µPa•s	µPa•s	µPa•s	µPa•s	µPa•s	$\sigma$ (weighted)
298.15	4	11.83	$17.788 \pm 0.002$	$3.150\pm0.042$	$8.624 \pm 0.198$	$-4.171 \pm 0.317$	$2.474 \pm 0.160$	0.037
323.15	4	10.90	$18.914 \pm 0.004$	$3.264 \pm 0.067$	$7.662 \pm 0.322$	$-2.652 \pm 0.538$	$1.781 \pm 0.287$	0.041
348.15	4	10.06	$20.010 \pm 0.003$	$3.095 \pm 0.050$	$8.046 \pm 0.258$	$-3.155 \pm 0.464$	$2.021 \pm 0.267$	0.027
373.15	3	9.37	$21.041 \pm 0.003$	$3.351 \pm 0.042$	$6.233 \pm 0.129$	$0.328 \pm 0.107$		0.039
398.15	3	8.83	$22.060 \pm 0.003$	$3.234\pm0.037$	$6.311 \pm 0.120$	$0.311\pm0.106$		0.031
423.15	2	8.32	$23.049\pm0.002$	$3.049 \pm 0.012$	$6.715\pm0.017$			0.020

Table 3. Coefficients of Equation 2

	$\eta_{i0}$	$\eta_{i1}$	$\eta_{i2}$	
i	µPa•s	µPa•s	µPa•s	$\sigma$ (weighed)
0	$46.518 \pm 0.075$	$-104.68 \pm 0.418$	$86.983 \pm 0.573$	
1	$0 \pm 0$	$16.471 \pm 0.283$	$-21.257 \pm 0.887$	
2	$9.922\pm0.305$	$-21.603 \pm 1.748$	$43.360 \pm 3.536$	
3	$0 \pm 0$	$0\pm 0$	$-22.240 \pm 2.175$	
4	$0\pm 0$	$55.481 \pm 0.460$	$0\pm 0$	0.057

isotherm of Gracki et al. measured up to densities of  $9.4 \text{ mol}\cdot\text{dm}^{-3}$  are systematically lower by approximately 0.4 %.

Evers et al.<sup>28</sup> carried out absolute measurements with their rotating-cylinder viscometer at densities up to 10.7 mol·dm<sup>-3</sup> in a large temperature range between (233 and 523) K. The comparison of the data by Evers et al. with the present values in the overlapping temperature ranges is illustrated in Figure 10. The deviations exceed  $\pm$  0.4 % only distinctly for the highest density at 293 K and at the highest common temperature of 423 K for the complete density range. The disagreement at 423 K needs further investigation in both laboratories.

#### Conclusions

The viscosity measurements of the present paper performed on nitrogen with a vibrating-wire viscometer are generally characterized by a reproducibility of  $\pm$  0.05 % and an uncertainty of  $\pm$  (0.2 to 0.3) %. The experimental values of the six isotherms were separately evaluated in terms of the reduced density using a polynomial series expansion with a maximum power of fourth order. In addition for the purpose of comparison, the values of all isotherms together were fitted with a double polynomial series expansion in terms of the reduced density and of the reciprocal reduced temperature.

The comparison with the most recent correlation of Lemmon and Jacobsen<sup>4</sup> makes evident that the uncertainties estimated



**Figure 6.** Comparison of experimental viscosity data from the literature with the results of the present paper at higher densitites. Deviations [ $\Delta = 100(\eta_{exp} - \eta_{present})/\eta_{present}$ ] as a function of molar density ( $\rho$ ). Experimental data:  $\bigcirc$ , Kestin and Leidenfrost,<sup>14</sup> (293–298) K;  $\triangle$ , Kestin and Leidenfrost,<sup>31</sup> (293–296) K;  $\diamondsuit$ , Kestin et al.,<sup>32</sup> 298 K.

for this correlation are too large and can be further improved. This needs a critical judgment of the techniques and the associated theories applied for the measurements as well as a mutual comparison of the high-quality data. Thus the most reliable data<sup>19,23–25,28</sup> including those of the present paper demonstrate that an improved correlation could be characterized in the dilute-gas region by uncertainties of  $\pm$  (0.1 to 0.2) % between room temperature and 420 K, possibly even up to 650 K. The comparison with experimental data, measured by Kestin et al.,<sup>14,31,32</sup> Timrot et al.,<sup>23</sup> Hoogland et al.,<sup>19</sup> Hurly et al.,<sup>29</sup> Flynn et al.,<sup>38</sup> Gracki et al.,<sup>39</sup> and Evers et al.,<sup>28</sup> and the critical



**Figure 7.** Comparison of experimental viscosity data from the literature with the results of the present paper at higher densitites. Deviations [ $\Delta = 100(\eta_{exp} - \eta_{present})/\eta_{present}$ ] as a function of molar density ( $\rho$ ). Experimental data:  $\odot$ , Timrot et al.,<sup>23</sup> (295–298) K;  $\otimes$ , Timrot et al.,<sup>23</sup> 305 K;  $\boxplus$ , Timrot et al.,<sup>23</sup> (384–389) K;  $\bigcirc$ , Hoogland et al.,<sup>19</sup> 298 K;  $\triangle$ , Hoogland et al.,<sup>19</sup> 308 K;  $\diamondsuit$ , Hoogland et al.,<sup>19</sup> 318 K;  $\bigtriangledown$ , Hoogland et al.,<sup>19</sup> 323 K;  $\square$ , Hoogland et al.,<sup>19</sup> 333 K;  $\times$ , Hurly et al.,<sup>29</sup> 298 K.



**Figure 8.** Comparison of the experimental viscosity data of Michels and Gibson<sup>37</sup> with the results of the present paper at higher densities. Deviations  $[\Delta = 100(\eta_{exp} - \eta_{present})/\eta_{present}]$  as a function of molar density ( $\rho$ ). Experimental data:  $\bigcirc$ , 298 K;  $\triangle$ , 323 K;  $\diamondsuit$ , 348 K.



**Figure 9.** Comparison of experimental viscosity data from the literature with the results of the present paper at higher densitites. Deviations [ $\Delta = 100(\eta_{exp} - \eta_{present})/\eta_{present}$ ] as a function of molar density ( $\rho$ ). Experimental data:  $\bigcirc$ , Flynn et al.,<sup>38</sup> 298 K;  $\triangle$ , Flynn et al.,<sup>38</sup> 373 K;  $\diamondsuit$ , Gracki et al.,<sup>39</sup> 298 K.



**Figure 10.** Comparison of the experimental viscosity data of Evers et al.<sup>28</sup> with the results of the present paper at higher densitites. Deviations [ $\Delta = 100(\eta_{exp} - \eta_{present})/\eta_{present}$ ] as a function of molar density ( $\rho$ ). Experimental data:  $\bigcirc$ , 293 K;  $\triangle$ , 333 K;  $\diamondsuit$ , 373 K;  $\bigtriangledown$ , 423 K.

discussion of the corresponding experiments lead to the conclusion that the uncertainties of a correlation based on these data could be decreased to  $\pm$  0.4 % in the density range up to about 10 mol·dm<sup>-3</sup> between ambient temperature and 400 K. Considering the differences to the results of the present work and the data by Evers et al., there is a need for further research at high densities and at temperatures above 400 K.

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